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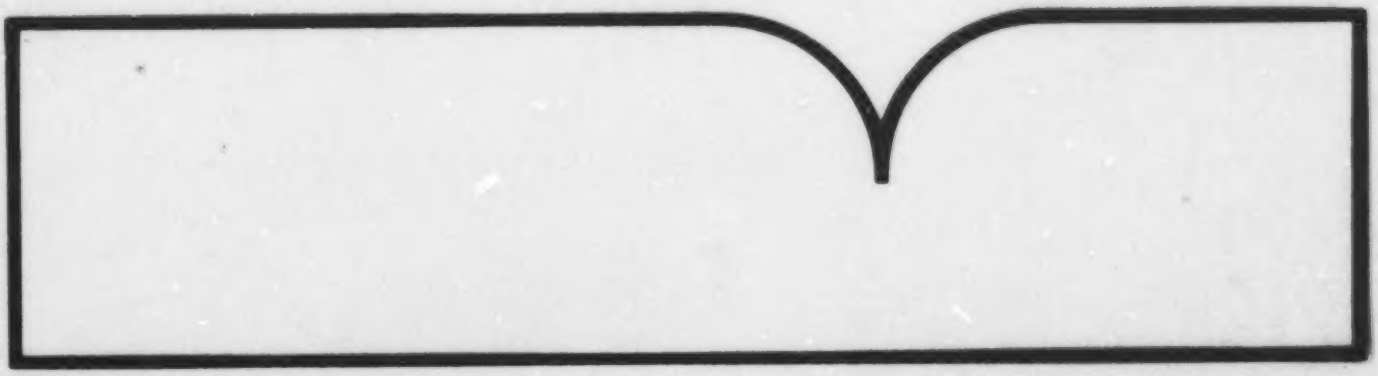

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16. ABSTRACT
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This paper is an attempt to illuminate some of these questions. The approach will be to review various steps in the transformation of the concepts of acid precipitation and atmospheric deposition from the domain of scientific curiosity to the domain of public concern and debate about energy and environmental policy.

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CHAPTER 3

AN HISTORICAL RESUME OF PROGRESS IN SCIENTIFIC AND PUBLIC UNDERSTANDING OF ACID PRECIPITATION AND ITS BIOLOGICAL CONSEQUENCES

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INTRODUCTION

Some years ago the terms "acid precipitation" and "acid rain" were bits of esoteric jargon used almost exclusively by scientists in certain specialized fields of ecology and atmospheric chemistry. During the last few years, these terms have become worrisome household words in many countries around the world. How did this transition come about? Who was responsible? Why did it take so long for acid precipitation to be recognized as an important ecological and environmental problem? What factors of scientific awareness and public perception have influenced the course and development of research on acid precipitation and its biological effects?

This chapter is an attempt to illuminate some of these questions. Our approach will be to review various steps in the transformation of the concepts of acid precipitation and atmospheric deposition from the domain of scientific curiosity to the domain of public concern and debate about energy and environmental policy.

Table 1 contains an historical resume of progress in understanding the phenomenon and its biological consequences. Contributions are presented in the chronological order of their occurrence whether or not the work was recognized or accepted at the time. Names of major contributors, the country in which the research was achieved, and the principal contribution to science or public affairs are presented with appropriate references when this is possible.

IN: Acid Precipitation Effects on Ecological Systems, F. M. D'Itri, ed., Chap. 3, pp. 43-83. Ann Arbor Science Publishers, Ann Arbor, MI, 1982

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Table 1

An Historical Resume of Progress in Understanding Acid Precipitation and Its Consequences

Year(s)	Investigator and Country	Principal Contribution to Understanding
1661-1662	Evelyn, Graunt, England	Noted the influence of industrial emissions on the health of plants and people, the transboundary exchange of pollutants between England and France, and suggested remedial measures including placement of industry outside of towns and use of taller chimneys to spread the "smoke" into "distant parts" (Evelyn, 1661; Graunt, 1662; see also Gorham, 1980).
1687	Hooke, England	On the basis of experiments by Brotherton, Hooke concluded that plants have a "two-fold kind of roots, one that branches and spreads into the earth, and another that spreads and shoots into the air, both kinds of roots serve to receive and carry their proper nourishment to the body of the plant." (Hooke, 1687; see also Gorham, 1965; and Wittwer and Bukovac, 1969).
1727	Hales, England	Noted that dew and rain "contain salt, sulphur, etc. For the air is full of acid and sulphureous particles..." (Hales, 1727; see also Gorham, 1980).
1734	Linné, Sweden	Described a 500-year-old smelter at Falun, Sweden: "...we felt a strong smell of sulphur,...rising to the west of the city...a poisonous, pungent sulphur smoke, poisoning the air wide around...corrode(ing) the earth so that no herbs can grow around it." (Linné, 1734).

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
1852	Smith, England	Analyzed the chemistry of rain near Manchester, England, and noted concentric zones with "three kinds of air--that with carbonate of ammonia in the fields at a distance,--that with sulphate of ammonia in the suburbs,--and that with sulphuric acid, or acid sulphate in the town." Smith also noted that sulfuric acid in town air caused fading in the color of textiles and corrosion of metals (Smith, 1852, see also Gorham, 1980).
1854-1856	Austria & Germany	Established "General Citizens Laws" prohibiting disposal of wastes by individuals on a neighbor's property. In the interest of encouraging industrialization, however, these same laws specifically excluded industries from legal liability in cases of waste disposal by pollution of water and air.
1855-1856	Way, England	Completed a very detailed series of analyses of nutrient substances in precipitation at the Rothamstead Experiment Station and showed the value of these substances in crop production (Way, 1855).
1872	Smith, England	In a remarkable publication entitled "Air and Rain: The Beginnings of A Chemical Climatology," Smith first used the term "acid rain" and enunciated many of the ideas that we now consider part of the acid precipitation problem. These ideas included regional variation in precipitation chemistry as it is influenced by such factors as combustion of coal,

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
		decomposition of organic matter, wind direction, proximity to the sea, amount of rain, etc. After extensive field experiments, Smith proposed detailed procedures for the proper collection and chemical analysis of precipitation. He also noted acid rain damage to plants and materials and atmospheric deposition of arsenic, copper, and other metals in industrial regions (Smith, 1872; see Gorham, 1980).
1881	Brögger, Norway	Observed "smudsig snefeld" (dirty snowfall) in Norway and attributed it to either a large town or an industrial district in Great Britain (Brögger, 1881).
1909	Sørensen, Denmark	Developed the pH scale to describe the acidity of aqueous solutions (Sørensen, 1909).
1911	Crowther & Ruston, England	Demonstrated gradients in acidity of precipitation decreasing from the center of Leeds, England, associated the acidity with combustion of coal, and showed that both natural rain and dilute sulfuric acid inhibited plant growth, seed germination, and ammonification, nitrification, and nitrogen fixation in soil (Crowther and Ruston, 1911; see also Cohen and Ruston, 1912; and Gorham, 1980).
1919	Rusnov, Austria	Demonstrated that deposition of substances from the atmosphere accelerated the acidification of both poorly-buffered and well-buffered forest soils (Rusnov, 1919).

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
1921	Dahl, Norway	Recognized the relationship between acidity of surface water and production of trout (Dahl,1921; 1927).
1922	Atkins, England	Measured the alkalinity of surface waters and noted a relationship between alkalinity and biological productivity (Atkins, 1922).
1923	MacIntyre, USA	Made the first detailed study of precipitation chemistry in the United States (MacIntyre and Young, 1923).
1925	Shutt & Hedley, Canada	Made very early measurements of the nitrogen compounds present in rain and snow and commented on the value of these compounds for growth of crops (Shutt and Hedley, 1925).
1926	Sunde, Norway	Demonstrated the value of adding limestone to water in a fish hatchery (Sunde, 1926).
1939	Erichsen-Jones, Sweden	Demonstrated the relationship between acidity and the toxicity of aluminum to fish (Erichsen-Jones, 1939).
1939	Katz et al., Canada	Reported acidification and decrease in base saturation of soils by sulfur dioxide emissions from the lead-zinc smelter near Trail, British Columbia (Katz et al., 1939).
1939	Bottini, Italy	Detected hydrochloric acid in precipitation near the volcano on Mount Vesuvius, thus demonstrating that there are natural sources of strong acids in precipitation (Bottini, 1939).

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
1942	Conway, Ireland	Completed the first modern review of precipitation chemistry (Conway, 1942).
1948	Egnér, Sweden	Initiated the first large-scale precipitation chemistry network in Europe (Egner et al., 1955; Rossby and Egner, 1955).
1950-1955	Eriksson, Sweden	Enunciated a general theory of biogeochemical circulation of matter through the atmosphere (Eriksson, 1952; 1954; 1959; 1960).
		Expanded the regional network established by Egnér into the continent-wide European Air Chemistry Network which has provided a continuing record of precipitation chemistry for 3 decades (Emanuelsson et al., 1954).
1953	Viro, Finland	Developed a regional chemical budget by comparing analytical data for precipitation and river waters in Finland (Viro, 1953).
1953-1958	Tamm, Sweden	Demonstrated the great dependence of mosses on atmospheric sources of nutrients, especially nitrogen (Tamm, 1953) and expanded this concept to include most forest plants (Tamm, 1958).
1953-1955	Various investigators in several countries	Simultaneously investigated precipitation chemistry data for evidence of atmospheric acidity (Barret and Brodin, 1955 - Sweden; Parker, 1955, and Gorham, 1955 - England; Houghton, 1955 - USA).
1954-1961	Gorham, England	Demonstrated that acidity in precipitation markedly influenced

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
		geological weathering processes and the chemistry of lake waters, bog waters, and soils (Gorham, 1955; 1958b, 1958c, 1961).
		Demonstrated that hydrochloric acid from combustion of coal rich in chlorine predominated in urban precipitation whereas sulfuric acid predominated in rural precipitation (Gorham, 1958a, 1958b).
		Established that acidity in precipitation affects the alkalinity and buffering capacity of lake and bog waters (Gorham, 1957; 1958b).
		Established that the incidence of bronchitis in man can be correlated with the acidity of precipitation (Gorham, 1959).
1957	Europe, USSR & USA	During the International Geophysical Year, a one-year study of precipitation chemistry was made in Europe, the USSR, and the USA.
1958	Junge & Werby, Jordan et al., USA	Made the first regional studies of precipitation chemistry in the United States and noted the importance of atmospheric sulfur as a source of nutrients for crops (Junge and Werby, 1958; Jordan et al., 1959).
1959	Dannevig, Norway	Recognized the relationship between acid precipitation, acidity in surface waters, and disappearance of fish (Dannevig, 1959).
1960-1963	Gordon & Gorham, Canada	Established that fumigation by sulfur dioxide and resultant acid rain contributed to the deterioration of

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
		lake quality, vegetation, and soils near metal smelters (Gorham and Gordon, 1960; Gordon and Gorham, 1963).
1960-1967	McCormick, USA	The first continent-wide precipitation chemistry network in North America was operated for 6 years -- 1960-66. The data showed that precipitation is generally acidic east but alkaline west of the Mississippi River (Lodge et al., 1968).
1962	Carson, USA	Stimulated a global revolution in environmental awareness by publishing "Silent Spring" in which the term "poison rain" was used to describe concern about atmospheric transport and deposition of pollutants (Carson, 1962).
1967-1968	Odén, Sweden	Outlined the changing acidity of precipitation as a regional and temporal phenomenon in Europe (Odén, 1968; 1976).
		Used trajectory analysis of air masses to demonstrate that acidity in precipitation in Scandinavia was attributable largely to emissions of sulfur in England and central Europe (Odén,1968).
		Demonstrated temporal trends in acidity and major cations and anions in precipitation in various parts of Europe (Odén, 1968).
		Demonstrated the increasing acidity of Scandinavian rivers (Odén, 1968).

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
		Described biological uptake and ion-exchange processes by which natural acidification of soils would be accelerated by atmospheric deposition of ammonia and other cations (Odén, 1968).
		Postulated that acid precipitation would lead to displacement of nutrient cations, reduction in nitrogen fixation, and release of heavy metals (especially Hg) which would damage surface waters and ground waters (Odén, 1968; 1976).
		Postulated acidity in precipitation as a probable cause of decline in fish populations, impoverishment of forest soils, decreased forest growth, increased disease in plants, and accelerated corrosion and other damage to materials (Odén, 1967; 1968).
1970	Odén & Ahl, Sweden	Discovered that soluble pollutants in snow accumulate in the snow pack and are released almost totally with the first meltwater in spring (Odén and Ahl, 1970).
1970	Hultberg, Sweden	Demonstrated the effect of acidity on fish populations in two Swedish lakes (Hultberg and Stenson, 1970).
1972	Bolin, Sweden	Drafted Sweden's Case Study for the United Nations Conference on the Human Environment "Air Pollution Across National Boundaries: The Impact on the Environment of Sulfur in Air and Precipitation". Noted damage to materials as well as ecosystems by acid precipitation (Bolin et al., 1972).

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
1972	Hvatum, Norway	Demonstrated increasing content of lead near the surface of Norwegian peat bogs and postulated long-distance transport as the probable cause (Hvatum,1972).
1972	Tyler, Sweden	Reported heavy metal accumulation in forest soils and speculated on their probable effects on forest productivity (Tyler,1972).
1972	Granat, Sweden	Described the temporal and spatial deposition of sulfate and acid over northern Europe (Granat, 1972; 1978).
1972	Jonsson & Sundberg, Sweden	Established an experimental basis for the suspicion that acid precipitation had decreased the growth of forests in Sweden (Jonsson and Sundberg, 1972).
1972	Various Agencies, Norway	Initiated the SNSF Project "Acid Precipitation: Effects on Forests and Fish (see Ambio, 1976 and Braekke, 1976).
1972-1980	Overrein, Norway	Demonstrated accelerated loss of calcium and other cations from soils receiving acid precipitation (Overrein, 1972). Provided leadership for the SNSF Project from its founding in 1972 until its completion in 1980 (Ambio, 1976; SNSF,1980).
1972	Likens et al., USA	Discussed the regional distribution of acid precipitation and its significance for aquatic and terrestrial ecosystems in North America (Likens et al., 1972).

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
		Indicated that nitric acid resulting from atmospheric transformation of NO_x adds to the acidity of precipitation in the eastern United States (Likens et al., 1972).
1972	Jensen & Snekvik, Norway	Showed that acidity in lakes and streams caused major decreases in salmon and trout populations in Norway (Jensen and Snekvik, 1972).
1972	Beamish & Harvey, Canada	Reported decline in fish populations due to acidification of lake waters in Canada (Beamish and Harvey, 1972).
1972-1980	Various Investigators	Showed that mushrooms, mosses, and other vegetation in forests accumulate heavy metals, especially lead and cadmium. Wildlife feeding on these plants also accumulate the metals, sometimes making both the plants and the wildlife hazardous for human consumption (Munshower, 1972; Huckabee and Blaylock,1974; Tyler, 1980).
1973	Malmer, Sweden	Summarized research on the ecological effects of increasing sulfur deposition, especially with reference to Swedish conditions (Malmer, 1973).
1973	Dickson et al., Sweden	Reported on the pH status of 314 lakes in western Sweden (Dickson et al.,1973).
1973	Wiklander, Sweden	Proposed a general theory to account for the effects of acid precipitation on soil chemical properties (Wiklander, 1973).

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
1973-1977	Ottar, Norway	Provided leadership for a continent-wide study of the long-range transport of air pollutants by the Organization for Economic Cooperation and Development in Europe (Ottar, 1976; OECD, 1977).
1974	Almer et al., Sweden	Summarized the effects of changing acidity of lake waters on fish populations in Sweden (Almer et al., 1974).
1974	Grahn et al., Sweden	Discovered that Sphagnum (peat moss) invades acidified lakes and streams, inducing a self-accelerating oligotrophication of the water body (Grahn et al., 1974).
1974	Cogbill & Likens, USA	Published maps showing changes in acidity of precipitation in the eastern United States between 1955-56 and 1972-73 (Cogbill and Likens, 1974).
1974	Hutchinson & Whitby, Canada	Established that strongly acid rain near Sudbury, Ontario, is accompanied by deposition and/or mobilization of heavy metals (especially nickel, copper, cobalt, iron, aluminium, and manganese). The toxicity of these metals was sufficient to inhibit germination and establishment of many native and agricultural species of plants (Hutchinson and Whitby, 1974; Whitby and Hutchinson, 1974).
1974	Shriner, USA	Demonstrated that simulated rain acidified with sulfuric acid can accelerate erosion of protective waxes on leaves, inhibit nodulation of leguminous plants, and alter

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
		host-pathogen interactions of plants (Shriner, 1974; 1976; 1978).
1975	Brosset et al., Sweden	Described the chemical transformations and trajectories that lead to "white episodes" and "black episodes" of acid deposition in western Sweden. Showed that gaseous nitric acid increases acid deposition. Established a state-of-the-art titration method for the determination of acidity in precipitation (Brosset et al.,1975).
1975-1980	Cowling, USA	Testified in Congressional hearings concerning the inadequacy of research in the U.S. on the ecological effects of acid precipitation (Cowling,1976).
		Together with many other scientists in the U.S. and Canada, began the development of a permanent network to monitor chemical changes in wet and dry deposition and to study their biological effects in various regions of the United States (Galloway and Cowling, 1978).
1975	Cragin et al., Greenland	Determined the chemistry of precipitation in Greenland from 1300 to 1975 A.D. The data show a continuing trend of increasing sulfate and lead concentration beginning about 1800 and rising even more rapidly after 1945 (Cragin et al.,1975).
1976	Schofield, USA	Reported the results of lake surveys showing decline in fish populations associated with acidification of lakes in the Adirondack Mountains of New York State (Schofield, 1976).

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
1976	Kucera, Sweden	Summarized evidence that acid precipitation accelerates the corrosion of metals (Kucera, 1976).
1976	Summers & Whelpdale, Canada	Summarized earlier studies of precipitation chemistry in Canada and identified northwest Alberta, southern Ontario and Quebec, and the Atlantic Provinces as areas of present or potential impact by acid rain and snow (Summers and Whelpdale, 1976).
1976	Leivistad & Muniz, Norway	Documented a massive fish kill on the Tovdal River associated with snow melting in the spring of 1975 and established that dead and dying fish had lost control of their blood salt balance (Leivistad and Muniz, 1976; Leivistad et al., 1976).
1976	Tyler, Sweden	Demonstrated that heavy metals in the litter layer of forests inhibit microbial processes, especially decomposition of organic matter (Tyler, 1976).
1976	Galloway, USA Berry, Canada Granat, Sweden	Developed standardized protocols for precipitation collectors and collection techniques (Galloway and Likens, 1976; Berry et al., 1976; Granat, 1976).
1976	Hultberg & Grahn, Sweden	Discovered a correlation between acidification of lakes and the mercury content of fish (Hultberg and Grahn, 1976).
1976	Pough, USA	Showed that reproduction of salamanders is inhibited by acidity of surface waters (Pough, 1976).

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
1977	Hagström, Sweden	Showed that reproduction of frogs is inhibited by acidity of surface waters (Hagström, 1977).
1977	Rosenqvist, Norway	Enunciated a general theory that acidification of soils and surface waters is due mainly to natural processes in soils and to changes in patterns of land use (Rosenqvist, 1977).
1978	Greszta, Austria	Demonstrated accumulation of heavy metals in forest soils leading to injury to young pine and spruce seedlings (Greszta, 1978).
1978-1980	Several Investigators, USA	Showed that acid precipitation occurs in both urban and certain rural areas in the western United States (Liljestrand and Morgan, 1978; McColl and Bush, 1978; Lewis and Grant, 1980).
1979	Altshuller, USA; McBean, Canada	Documented the transboundary exchange of sulfur and nitrogen between the United States and Canada (Altshuller and McBean, 1979c, 1980).
1979	Odén, Sweden	Showed by studies of surface waters in Sweden that acidification due to sulfur deposoition had begun during the early 1900's (Odén, 1979).
1979	Liljestrand & Morgan, USA	Completed a rigorous statistical analysis of trends in precipitation chemistry in the eastern and western United States (Liljestrand and Morgan, 1979).
1979	Cronan & Schofield, USA	Discovered that aluminum ions are leached by acid precipitation from soils into streams and lakes in

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
		concentrations toxic to fish (Cronan and Schofield, 1979).
1979	Carter, USA	Established a Presidential initiative on acid rain calling for a 10 year long, $10 million per year interagency program of research on acid precipitation and its environmental consequences in the USA (Carter, 1979).
1979	Henriksen, Norway	Developed a simple descriptive model for determining the extent to which acidification decreases the alkalinity of lake waters and applied this method to predict the vulnerability of lakes to acid deposition (Henriksen, 1979).
1980	Hultberg & Wenblad, Sweden	Discovered acidification of ground water in western Sweden, postulated acid precipitation as the probable cause, established by surveys of 1300 wells the frequency of heavy metal accumulation and plumbing problems associated with these wells (Hultberg and Wenblad, 1980).
1980	Abrahamsen, Norway	Summarized many years of research on the effects of acid precipitation on forests and concluded that fertilization effects, particularly by atmospheric deposition of nitrogen, tend to offset nutrient leaching and other detrimental effects. Also emphasized that negative effects of atmospheric deposition on growth are most likely when nutrient deficiencies or imbalances are increased by acid deposition (Abrahamsen, 1980).

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
1980	Wetstone, USA	Summarized the biological and materials-damage effects of acid precipitation in relation to the pollution control laws of North America (Wetstone, 1980).
1980	Schindler et al., Canada	Established an experimental system for controlled acidification of whole lakes (Schindler et al., 1980).
		Demonstrated that acidification eliminated organisms of several trophic levels at pH values as high as 5.8 to 6.0 (Schindler, 1980). Also demonstrated that microbial reduction of sulfate could partially protect lakes against acidification (Schindler et al., 1980).
1980	Ulrich et al., Germany	Demonstrated a significant correlation between amount of soluble aluminum in forest soils, death of feeder roots in spruce, fir, and birch forests, and widespread decline in the growth of these forests (Ulrich et al., 1980; Schuck et al., 1979).
1980	Various Investigators, USA	Established an Integrated Lake-Water Acidification Study (ILWAS), to determine detailed chemical budgets for hydrogen, sulfate, ammonium, nitrate, chloride, and other ions in three lake-watersheds of differing degrees of acidification (see Drabløs and Tollan, 1980, pp. 252-267; and EPRI, 1981).

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
		MAJOR INTERNATIONAL CONFERENCES DOCUMENTING FURTHER PROGRESS
1950's	Eriksson, Sweden	The International Meteorological Institute sponsored a series of conferences on various aspects of atmospheric chemistry (Eriksson, 1954).
1975-1976	Dochinger & Seliga, USA	U.S. Forest Service sponsored the First International Symposium on Acid Precipitation and the Forest Ecosystem at the Ohio State University, Columbus, Ohio (Dochinger and Seliga, 1976a; 1976b).
1976	Overrein et al., Norway	SNSF Project and the Norwegian Ministry of the Environment sponsored the International Conference on Effects of Acid Precipitation, Telemark, Norway (Ambio, 1976; Braekke, 1976).
1977	Husar & Lodge, USA; Moore, England	The United Nations Environmental Program and several other organizations sponsored an International Symposium on Sulfur in the Atmosphere, Dubrovnik, Yugoslavia (Husar et al., 1978).
1978	Hutchinson, Canada	NATO sponsored an Advanced Research Institute on Ecological Effects of Acid Precipitation in Toronto, Canada (Hutchinson and Havas, 1980).
1978	Howells, England	The Electric Power Research Institute (USA) and the Central Electricity Generating Board (Great Britain) sponsored an International Symposium on Ecological Effects of Acid Precipitation (Howells, 1979).

Table 1 (continued)

Year(s)	Investigator and Country	Principal Contribution to Understanding
1978	Hendrey, USA	U.S. Environmental Protection Agency and Brookhaven National Laboratory sponsored an International Workshop on Limnological Aspects of Acid Precipitation, Sagamore Lake, New York (Hendrey, 1978).
1979	Shriner, USA	The Oak Ridge National Laboratory sponsored a symposium on the Potential Environmental and Health Effects of Sulfur Deposition, Gatlinburg, Tennessee (Shriner et al., 1980).
1979	Agle, USA & Turnbull, Canada	A group of Canadian and United States environmental protection organizations sponsored the Action Seminar on Acid Precipitation, Toronto, Canada (Reid, 1980).
1980	Overrein et al., Norway	The SNSF Project sponsored an International Conference on Ecological Impacts of Acid Precipitation, Sandefjord, Norway.
1980	Miller et al., USA	The U.S. Forest Service and other U.S. and international air pollution organizations sponsored an International Symposium on Effects of Air Pollutants on Mediterranean and Temperate Forest Ecosystems, Riverside, California.

Selecting particular contributions for inclusion in such a chronology is a hazardous business because of the likelihood that some important contributions will not be given the credit and/or priority they deserve. There is no choice but to accept the blame for whatever errors of omission and ignorance are evident in Table 1. I hope the Table will serve a useful purpose,

however, and look forward to receiving comments from any person who may be interested to help describe the record more adequately.

EARLY AWARENESS OF ACID PRECIPITATION

Many features of the acid rain phenomenon were first discovered by an English chemist named Robert Angus Smith in the middle of the 19th century. In 1852, Smith published a detailed report on the chemistry of rain in and around the city of Manchester in England. In this account, Smith (1852) called attention to the changes in precipitation chemistry as one moves from the middle of a polluted city to its surrounding countryside: "We may, therefore, find three kinds of air--that with carbonate of ammonia in the fields at a distance; that with sulphate of ammonia in the suburbs; and that with sulphuric acid, or acid sulphate, in the town." Smith also pointed out that the sulfuric acid in city air caused the fading of colors in textiles and the corrosion of metals.

Twenty years later, in a remarkable book entitled "Air and Rain: The Beginnings of A Chemical Climatology," Smith (1872) first used the term "acid rain" and enunciated many of the principal ideas that are part of our present understanding of the acid rain phenomenon. On the basis of detailed studies in England, Scotland, and Germany, Smith demonstrated that precipitation chemistry is influenced by such regional factors as combustion of coal, decomposition of organic matter, wind trajectories, proximity to the sea, and amount and frequency of rain or snow. Smith proposed detailed procedures for the proper collection and chemical analysis of precipitation. He also noted acid rain damage to plants and materials in industrial regions.

Unfortunately, however, Smith's pioneering and prophetic book apparently has been overlooked by essentially every subsequent investigator of the acid precipitation phenomenon. Eville Gorham (1980) has developed the first detailed analysis of Smith's early work for a report by the National Academy of Sciences.

MODERN AWARENESS OF ACID PRECIPITATION

Modern understanding of acid precipitation and its environmental consequences had its origins in three seemingly unrelated fields of science: limnology (the study of

freshwaters), agriculture, and atmospheric chemistry. Let us look at the contribution of each field of science in turn.

Progress in Limnology

The relationship between rain or snow, and the water in streams and lakes was obvious even to prehistoric man. But the relationship between changes in the chemistry of rain and snow and changes in the chemistry of lake waters remained obscure until the middle of the twentieth century.

During the first half of this century, several scientists in Europe and North America studied certain isolated aspects of atmospheric and surface water chemistry. Occasionally, interactions were demonstrated, but it was Eville Gorham, now Professor of Ecology of the University of Minnesota, who, in a long series of papers, developed the major foundations for our present understanding of the causes of acidity in precipitation and its impact on aquatic ecosystems (Gorham, 1955; 1957, 1958a-c; 1959; 1961; 1965). On the basis of research both in England and in Canada, Gorham and his colleagues demonstrated the following principles:

(1) Much of the acidity of precipitation in industrial regions can be attributed to atmospheric emissions produced during combustion of fossil fuels.
(2) Progressive loss in alkalinity of surface waters and increase in the acidity of bog waters can be traced to atmospheric deposition of acidic substances in precipitation.
(3) Much of the free acidity in soils receiving acid precipitation is due to sulfuric acid.
(4) The incidence of bronchitis in man can be correlated with the acidity of precipitation.
(5) Fumigation with sulfur dioxide and resultant acid rain contribute to the deterioration of vegetation, soils, and lake water quality around metal smelters.

Thus, by the mid 1950s and early 1960s, Gorham had established a major part of our present understanding of the sources and the limnological and ecological significance of acid precipitation. But these pioneering researchers, like those of Smith a century before, were met by a thundering silence from both the scientific community and the public at large. Lack of recognition of the environmental significance of Gorham's work resulted in a further lag in both scientific and public awareness of acid precipitation and its ecological significance.

Progress in the Agricultural Sciences

The importance of the atmosphere as a source of nutrients for the growth and development of plants was first recognized by Robert Hooke in 1687 (see also Gorham, 1965). Early experiments at the Rothamsted Experiment Station in England also demonstrated the relationship between nutrients in air and the growth of various crop plants (Way, 1855). In the mid-1940s, an imaginative Swedish soil scientist, Hans Egnér, systematized investigations on the fertilization of crops by atmospheric sources of nutrients. Working from his laboratory at the Agricultural College near Uppsala, Egnér created the first twentieth century network for the collection and chemical analysis of precipitation and air at ground level. A large number of sampling buckets were set out at experimental farms all over Sweden and the major chemical constituents in what we now call bulk deposition (rain, snow, and dust fall) were measured on a regular monthly basis. The acidity of precipitation was one of several chemical parameters that were measured. This network, initiated by Egnér in 1948, was gradually spread by other agricultural scientists, first to Norway, Denmark, and Finland and later to most of western and central Europe. This network provided the first large scale and long-term data on the changing chemistry of precipitation and its importance for agriculture and forestry (Emanuelsson et al., 1954; Egnér et al., 1955; Tamm, 1958). For the first time it became possible to discern both temporal and spatial patterns of change in precipitation chemistry. In 1956, the International Meteorological Institute in Stockholm assumed responsibility for further coordination and expansion of Egnér's network into the European Air Chemistry Network. In 1957, as part of the International Geophysical Year, the European Network spread eastward to include Poland and much of the Soviet Union. In marked contrast to monitoring efforts elsewhere in the world, most stations of the original European Network, including those in Poland and the Soviet Union, have remained in continuous operation for nearly three decades!

Progress in the Atmospheric Sciences

Under the imaginative leadership of Carl Gustav Rossby and Erik Eriksson, the science of atmospheric chemistry began in Sweden and later spread across Europe and finally to North America (NAS, 1975). Rossby and Eriksson were convinced that atmospheric processes were efficient mechanisms for the long-distance as well as the short-distance dispersal of all sorts of substances. The data from Egnér's precipitation chemistry

network provided the means to test various hypotheses about the trajectories of air masses, turbulent dispersal processes, and atmospheric scavenging and deposition processes (Rossby and Egnér, 1955).

The transport and deposition of water by atmospheric processes have been well known since the earliest observations of evaporation, wind and cloud movement, and of rain and snow. But the notion that atmospheric transport and deposition processes were major means for dispersal and chemical transformation of many other substances was still only a working hypothesis in the early 1950s. Rossby and Eriksson championed these then novel ideas and initiated various experimental tests of their hypotheses. Data from the European Air Chemistry Network provided a very powerful tool for these studies. Rossby and Eriksson also sponsored a series of European conferences on atmospheric chemistry and dispersal processes. These conferences attracted the interest of scientists in many other fields including biology, forestry, agriculture, meteorology, engineering, and medicine (Eriksson, 1954).

Integration of Limnological, Agricultural, and Atmospheric Chemical Studies

The first major integration of knowledge about acid precipitation in these three fields of science was achieved by Svante Odén, a soil scientist at the Agricultural College near Uppsala in Sweden. As a young colleague of Egnér, Rossby, and Eriksson, Odén started a Scandinavian network for surface water chemistry in 1961. When data from this network were combined with those from the European Air Chemistry Network, a series of general trends and relationships began to emerge and were published by Odén in two different media: in Stockholm's prestigious newspaper Dagens Nyheter (Odén, 1967) and in an Ecology Committee Bulletin (Odén, 1968). The newspaper report outlining Odén's ideas about an insidious "chemical war" among the nations of Europe captured the attention of the press, who began the process of public education about acid precipitation in Europe. The Ecology Committee Bulletin had a similarly provocative influence on scientific interest in both the phenomenon and the ecological effects of the increasing acidity of precipitation. Suddenly, limnological, agricultural, and atmospheric scientists began talking to each other about Odén's unconventional ideas and the general theory of atmospheric influences which he enunciated. Odén's analyses of the trajectories and chemistry of air masses clearly showed that:

(1) acid precipitation was a large scale regional phenomenon in much of Europe with well defined source and sink regions;
(2) both precipitation and surface waters were becoming more acidic; and
(3) long-distance transport (100 to 2000 km) of both sulfur- and nitrogen-containing air pollutants was taking place among the various nations of Europe.

Odén also enunciated a series of hypotheses about the probable ecological consequences of acid precipitation with respect to:

(1) decline of fish populations;
(2) decreased forest growth;
(3) increased plant diseases; and
(4) accelerated damage to materials.

These conclusions and hypotheses led to a veritable storm of scientific and public concern about acid precipitation. The Swedish government responded to growing public and scientific concern by initiating an inquiry which culminated in Sweden's Case Study for the United Nations Conference on the Human Environment, "Air Pollution Across National Boundaries: The Impact of Sulfur in Air and Precipitation" (Bolin et al., 1972).

The major ideas in both the Ecology Committee Bulletin and the Swedish Case Study were much debated all over Europe. Two major scientific initiatives followed in short order. The first occurred in 1972 when three organizations in Norway joined together in establishing the so-called SNSF Project (The Norwegian Interdisciplinary Research Programme "Acid Precipitation: Effects on Forest and Fish"). These three organizations included the Norwegian Council for Scientific and Industrial Research, the Agricultural Research Council of Norway, and the Norwegian Ministry of Environment. The annual budget for the SNSF Project was about 10 million Norwegian Kroner (U.S., 2 million dollars) per year from 1972-1980. This huge project had two comprehensive goals:

(1) To establish as precisely as possible, the effects of acid precipitation on forests and freshwater fish.
(2) To investigate the effects of air pollutants on soils, vegetation, and water to the extent required to support the primary objective.

Lars Overrein of the Norwegian Forest Research Institute was appointed Research Director for this project and served in this capacity from the inception to the termination of the project in 1980. The project produced a steady stream of

technical and scientific reports on various aspects of acid precipitation and its effects. The SNSF Project also sponsored two major international scientific conferences, one at Telemark, Norway, in June 1976 and the second at Sandefjord, Norway, in March 1980. Braekke (1976) and Ambio (1976) published major reports in connection with the Conference at Telemark. The Conference at Sandefjord was designed to provide a forum for evaluation not only of recent research within the SNSF Project, but also that being done elsewhere in the world. A final report and bibliography for the SNSF Project has been published recently (SNSF, 1980).

The second major initiative was the so-called OECD Study which provided quantitative data on the long-distance transport and deposition of atmospheric sulfur in both eastern and western Europe. Brynjulf Ottar (1976) of the Norwegian Institute for Air Research provided leadership for this continent-wide study of the long-range transport of sulfur pollutants. The results, which were published in 1977 by the Organization for Economic Cooperation and Development (OECD, 1977), confirmed many of the ideas presented in the Ecology Committee Bulletin and the Swedish Case Study regarding the long-range transport and the exchange of sulfur dioxide and sulfuric acid among the nations of Europe.

Recent analyses of air mass movements in North America have shown that similarly massive long-range transport is taking place among the various states within the United States and between the United States and Canada.

Scientific and Public Awareness in North America

Concern about acid precipitation and its ecological effects in North America developed first in Canada and then later in the United States. Initial interests were focused on the effects of sulfur dioxide fumigation and associated acid precipitation and heavy metal deposition near metal smelting and sintering operations (Katz, 1939; Gordon and Gorham, 1963), especially those near Sudbury, Ontario, the largest point source of sulfur in the world (Gorham and Gordon, 1960; Hutchinson and Whitby, 1974). During the early 1970s, interest spread to other parts of Canada as declining fish populations were discovered in more and more lakes remote from local sources of atmospheric sulfur in southern Ontario and Nova Scotia (Beamish and Harvey, 1972). Very early measurements of nitrogen compounds in rain and snow were made by Shutt and Hedley (1925); the first measurements of the pH of precipitation in Canada

apparently were reported by Herman and Gorham (1957). The first national monitoring program for precipitation chemistry was initiated by the Canadian Department of the Environment in 1975. In recent years, several federal and provincial hearings on acid, sulfur, and heavy metal deposition have taken place in Canada as a result of growing public concern about acid precipitation. A summary of acid precipitation studies in Canada was published in 1976 by Summers and Whelpdale (1976).

The first detailed studies of precipitation chemistry in the United States were made by MacIntyre and Young (1923). This work was followed by the work of Junge and various collaborators during the 1950s (Junge and Werby, 1958). The first regional monitoring network for precipitation chemistry was maintained by a group of State Agricultural Experiment Station scientists during the period from 1953 to 1955 (Jordan et al., 1959). The first national monitoring program in the United States was established under the auspices of the Air Pollution Program within the Public Health Service Laboratory at Cincinnati, Ohio. The data for 1960 to 1966 were summarized by Lodge et al. (1968). As has been the case in all such studies in North America, these early programs were redirected and/or terminated so that no continuing records are available with which to evaluate long-term trends in the chemistry and acidity of precipitation.

Scientific and public interest in acid precipitation and its ecological consequences in North America were stimulated by Svante Oden in a series of 14 lectures at various institutions in the United States during the fall of 1971, and also by Torsten Ahl and Oden at the 19th International Limnological Congress in Winnipeg, Manitoba, in 1974. Further stimulus was provided by a series of publications by Gene Likens, Charles Cogbill, James Galloway, Carl Schofield, and others (Likens et al., 1972; Likens, 1976; Cogbill and Likens, 1974; Schofield, 1976; Galloway and Likens, 1976; Galloway et al., 1978; Likens et al., 1979). Experimental studies of various biological effects of acid precipitation were initiated at Cornell, North Carolina State, and other universities. David Shriner's (1976) dissertation demonstrated both direct injury to vegetation and various indirect effects through pathogens and parasites. Carl Schofield's (1976) research on extinction of fish populations in the Adirondack Mountains was especially alarming.

Growing awareness of important impacts of acid precipitation on fish populations and potential effects on forests led the U.S. Forest Service to sponsor the First International Symposium on Acid Precipitation and the Forest Ecosystem in Ohio in May 1975. The proceedings of this

symposium and the associated workshop report were published by Dochinger and Seliga (1976a, 1976b). At Congressional hearings in July 1975, Cowling (1976) testified on the inadequacy of research in the United States on acid precipitation. Specifically, lack of a coordinated program of research on ecological effects and lack of a stable monitoring network were recognized as primary causes of our profound ignorance of acid precipitation. In the spring of 1976, a cadre of many scientists in various institutions and agencies throughout the United States began the process of creating a National Atmospheric Deposition Program (NADP) to meet these two critical needs (CSRS, 1977; Kennedy, 1977; Galloway and Cowling, 1978).

In the fall of 1977, the President's Council on Environmental Quality contracted with the NADP for the drafting of "A National Program for Assessing the Problem of Atmospheric Deposition (Acid Rain)." This publication (Galloway et al., 1978) provided the basis for a Presidential Initiative on Acid Precipitation which President Carter announced on August 2, 1979, in his Second Environmental Message (Carter, 1979). This initiative calls for a 10 year long, 10 million dollars per year program of research on the causes and consequences of acid precipitation in the United States.

THE PRESENT STATUS OF KNOWLEDGE CONCERNING ACID PRECIPITATION

During the summer of 1979, a cooperative agreement was established between the U.S. Environmental Protection Agency, the National Atmospheric Deposition Program, and North Carolina State University for the management and coordination of research on the Effects of Acid Precipitation on Aquatic and Terrestrial Ecosystems. One of the major responsibilities specified in this agreement is synthesis and integration of knowledge about acid precipitation and its ecological effects. Accordingly, an effort was made to summarize in a few statements of fact the present status of knowledge in this area of science. The most recent version of this status report is presented by Cowling in Chapter 1 of this book. These summary statements are providing a part of the background upon which scientists in various institutions and organizations in the United States and Canada will continue to build a scientific foundation for understanding the phenomenon of acid precipitation and its important ecological effects.

THE FUTURE OF ACID PRECIPITATION RESEARCH

We have come a long way since the earliest attempts of Smith, Gorham, and Odén to alert the scientific community and the public at large to the causes and consequences of acid rain. Much has been learned both in Europe and in North America. But much more remains to be learned about many aspects of the phenomenon and its effects. The pathway that has led to our present understanding has been illuminated by the remarkable insight of a few imaginative scientists. It has also been illuminated by many others who have filled lesser voids in our knowledge.

Today, public interest in acid rain research is at an all time high in many parts of the world. The challenge for us as scientists is to satisfy that interest by providing a still deeper understanding of the atmospheric processes, the soils transformations, the vegetational changes, the alterations in water chemistry, the materials effects, the physiological influences, and the nutrient as well as toxic effects of acid precipitation.

Research is the key to improved understanding. Improved understanding is the key to wiser public and private decisions that relate to the use of energy and to the quality of life in our society.

Let us get on with the job of learning so that the challenge of managing acid precipitation and its effects can begin as soon as possible.

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